

An optical nanofiber–based interface for single molecules

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Optical interfaces for quantum emitters are a prerequisite for implementing quantum networks. Here, we couple single molecules to the guided modes of an optical nanofiber. The molecules are embedded within a crystal that provides photostability and, due to its inhomogeneous environment, a means to spectrally address single molecules. Single molecules are excited and detected solely via the nanofiber interface without the requirement of additional optical access. In this way, we realize a fully fiber–integrated system that is scalable and may become a versatile constituent for quantum hybrid systems.

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In recent years, single molecules in solids [1–7] and other solid state quantum emitters such as color centers in diamond [8–11] and quantum dots [12, 13] have gained increasing interest as building blocks for quantum networks [14, 15], quantum metrology [16–18] and nanosensors [19–21]. For all these applications a strong light–matter interaction is essential. This can be achieved by coupling to a large ensemble of quantum emitters [22, 23], by employing a cavity [24–27] or by decreasing the mode area of the interacting light field [28–31] and hence achieving a significant overlap between the absorption cross–section of the emitter and the respective light field. A versatile platform to achieve such a small mode area of the light field are optical nanofibers [31, 32]. An optical nanofiber is the waist of a tapered optical fiber (TOF) and has a diameter smaller than the wavelength of the light it is guiding. Therefore, an appreciable fraction of the light propagates outside the fiber in the form of an evanescent wave. Due to the strong transverse confinement of the light field, which prevails over the entire length of the nanofiber, the interaction with emitters close to the surface can be significant [30, 33–36].

Single molecules in crystalline solids are efficient quantum emitters that exhibit strong zero phonon lines (ZPL) which can be as narrow as tens of MHz at cryogenic temperatures [37, 38]. Due to inhomogeneous broadening caused by the host crystal such molecules can be spectrally discerned and individually addressed using a narrowband laser [39]. For a low concentration of molecules, this makes it possible to circumvent the additional spatial selection that has been used for numerous single molecule experiments in the past [40, 41], even if a large fraction of the crystal is illuminated. This was also exploited in a recent experiment by Faez et al. [28], where single dibenzoterrylene (DBT) molecules have been coupled to light propagating through a nanocapillary.

Here, we show for the first time that single organic molecules can be interfaced with an optical nanofiber.

This presents a new platform based on solid state emitters that can be used for quantum optics and is naturally integrated into optical fiber networks.

In our experimental set–up, the TOF resides inside a cryostat [Fig. 1(a)] and we interface terrylene molecules in a para–terphenyl (p–terphenyl) crystal with the evanescent light field surrounding its nanofiber. The latter has a total length of 3 mm and a diameter of 320 nm. The TOF is produced in a heat and pull process using a custom–made pulling rig [42]. In the 6.8 cm long tapered section of the fiber, the weakly guided LP₀₁ mode of the standard single mode optical fiber is adiabatically transformed into the strongly guided HE₁₁ mode of the nanofiber waist and back yielding transmission losses of less than 2% from 520–650 nm. For our purpose, a broadband transmission is crucial as the excitation and detection wavelengths can differ by more than 100 nm. This requires a careful choice of tapering angles and waist diameter [43]. Terrylene in p–terphenyl can exhibit four different electronic transition frequencies from the ground to the first excited state termed X1–X4, corresponding to four possible orientations of the molecules in the crystal. Molecules in the X4 orientation are resonant with light at 577.9 nm and have been shown to be very photostable [44]. Hence, all measurements presented here use molecules in this site. A simplified level diagram is depicted in Fig. 1(b). The laser excites the molecule on the zero phonon line 00ZPL that connects the ground and the excited electronic state without any vibrational contribution of the molecule. After excitation, the molecule will decay into any of the vibrational states in the electronic ground state with a probability determined by the Franck Condon and Debye–Waller factors. From these states it will nonradiatively decay into the vibrational ground state within picoseconds. Hence, the absorption cross–section [45] of a single molecule in a solid is

$$\sigma = \frac{3\lambda^2}{2\pi} \frac{\Gamma^2}{\Gamma_{\text{hom}}^2} \frac{(\mathbf{d} \cdot \mathbf{e})^2}{|\mathbf{d}|^2} \quad (1)$$

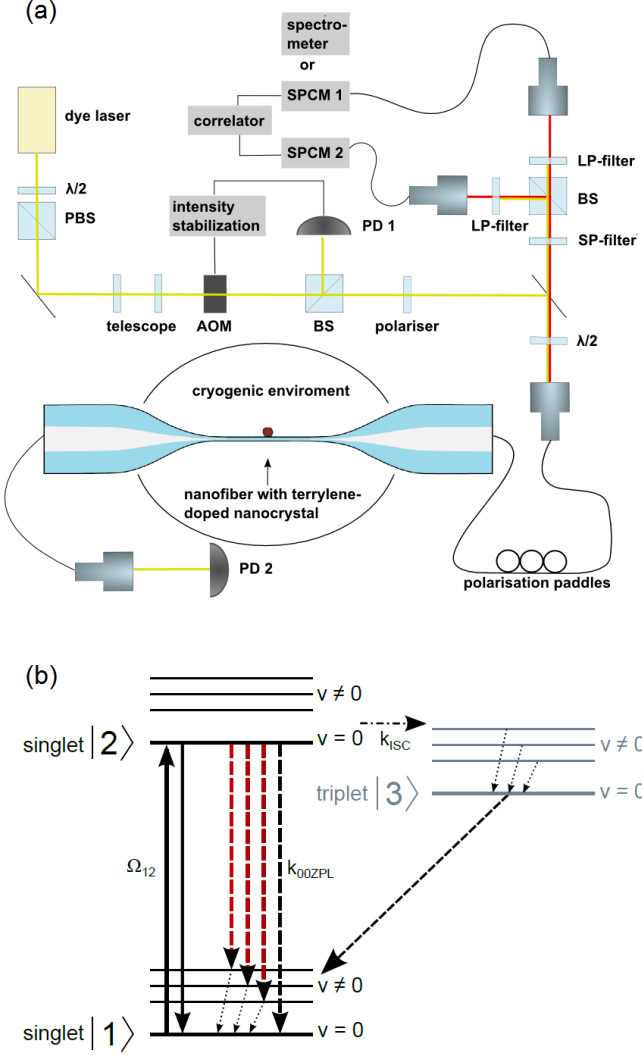


Figure 1. (a) Experimental set-up for single molecule spectroscopy via an optical nanofiber interface. (b) Simplified level diagram of terrylene in p-terphenyl. Full lines indicate transitions driven by the laser, while dashed lines represent spontaneous emission. The dot-dashed line marked k_{ISC} shows intersystem crossing from the excited singlet state to the triplet state. Dotted lines within an electronic manifold correspond to non-radiative decay processes that occur on a timescale of ps.

where the product $\mathbf{d} \cdot \mathbf{e}$ represents the projection of the polarization vector \mathbf{e} of the excitation light on the molecular dipole \mathbf{d} and λ denotes the excitation wavelength. Γ is the lifetime-limited linewidth and Γ_{hom} the homogeneously broadened linewidth of the molecule, respectively. If the dipole is aligned with the polarization of the excitation light and the homogeneous broadening is negligible, the absorption cross-section will be that of a simple two-level atom and is comparable to the effective mode area [46] of our optical nanofiber of about $0.5 \times \lambda^2$.

This ensures a strong effect of a single molecule on the light field. The probability of a terrylene molecule to decay to the triplet state after excitation rather than to the singlet ground state is very low and has experimentally been found to be $< 10^{-5}$ at cryogenic temperatures [2, 47]. To maintain sufficient light guiding capabilities of the nanofiber-crystal system for spectroscopy and to ensure that the crystal stays tightly adhered to the vertically mounted nanofiber, the crystals have to be on the order of a few hundred nanometers in size. Such nanocrystals are grown by a reprecipitation method [48] from an oversaturated solution of a 8×10^{-5} molar mixture of terrylene/p-terphenyl in toluene. The solution is heated until both compounds are dissolved and then isopropanol is added as a reprecipitation agent. This procedure results in terrylene doped p-terphenyl crystals of platelet morphology as seen in Fig. 2(a), which shows a scanning electron microscope (SEM) image of such crystals deposited on a silicon substrate. The majority of crystals have base dimensions in the range of 200 to 2000 nm and a base width to height ratio of 2.5:1 to 5:1 as determined by atomic force microscopy (AFM) measurements. The monocrystalline nature of these crystals has been verified by performing selected area electron diffraction (SAED) measurements using a transmission electron microscope (TEM), see Fig. 2(b). SAED also indicates that the substrate-supported crystal platelet base face is of (001) orientation, i.e. the crystal's c-axis is perpendicular to the substrate. It is known that the dipole moment of the transition to the lowest electronically excited state in terrylene is linear and lies along the long axis of the molecule [44]. When inserted into a p-terphenyl crystal, the molecule's long axis and thus the dipole moment lie nearly parallel to the crystal's c-axis and therefore roughly perpendicular to the substrate.

A terrylene-doped p-terphenyl nanocrystal is deposited on the nanofiber by a drop-touch method: a drop of the suspension of doped p-terphenyl nanocrystals is briefly brought into contact with the nanofiber via a pipette. During this process, the transmission of the excitation laser and the fluorescence of the nanofiber is monitored with a power meter and a spectrometer, respectively. When a doped crystal has adhered to the nanofiber surface during the brief contact with the suspension droplet, a typical fluorescence signal and some loss in transmission is observed. As crystals of a variety of sizes are produced during our growth process, we have to post-select the size of the deposited crystal. However, since the largest crystals sediment faster, we usually find suitably small ones in the suspension supernatant. If it is nevertheless found that the transmission deteriorates too much during the deposition process, the crystal can be washed off with acetone and another one deposited.

The TOF is mounted on a steel holder with two NdFeB magnets. This ensures that the fiber is firmly held and that it stays intact during the cooling process down to

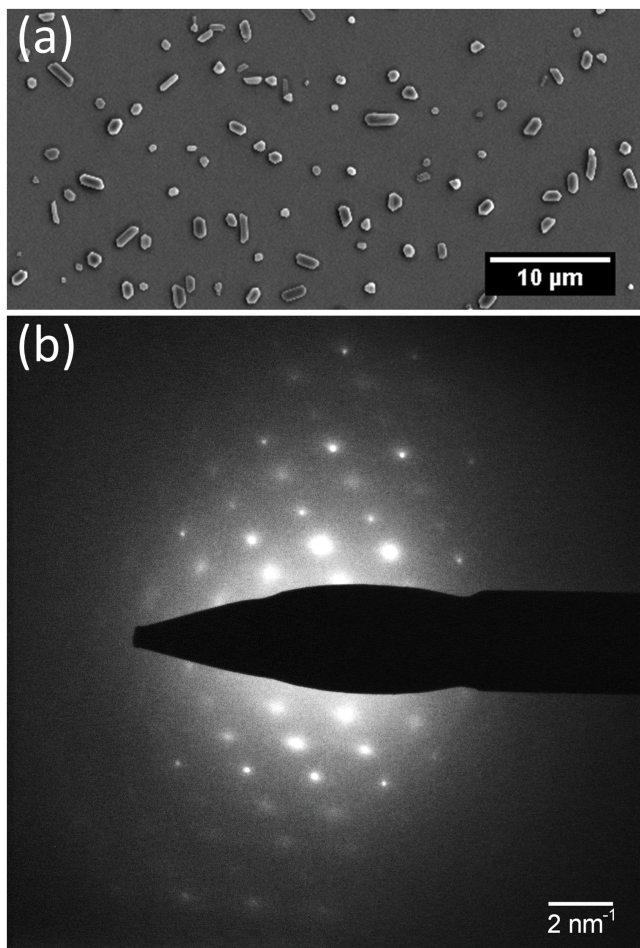


Figure 2. (a) Scanning electron microscope image (FEI Quanta 200 FEG) of terrylene-doped p-terphenyl crystals on a silicon substrate. (b) Selected area electron diffraction (SAED) pattern of an individual terrylene-doped p-terphenyl crystal (Philips CM200 TEM at 200 kV, imaged with the electron beam perpendicular to the crystal base face, exposure time was kept short (≤ 3 s) to avoid electron beam induced degradation of the organic crystal [49]). The pattern is consistent with the monoclinic high-temperature phase of a single p-terphenyl single-crystal [50] viewed along the [001] axis (considering double diffraction effects, Cambridge Structural Database (CSD) entry terphe14).

cryogenic temperatures. This fiber set-up is mounted in the cold-pot of a custom-made cryostat that can cool the sample to 4 K. To ensure efficient thermalisation of the nanofiber and the crystal with the walls of the cold-pot, helium buffer gas at a pressure of a few mbar is introduced into the cold-pot before cool-down, after it has been evacuated.

To excite the molecules, light of a Matisse dye laser is coupled into the optical fiber that connects to the TOF and that enters the cryostat via a teflon feed-through [Fig. 1(a)]. To compensate for intensity fluctuations and drifts, the laser beam is sent through an acousto-

optic modulator (AOM) and partly onto a photodiode (PD) and is actively intensity stabilised. A fraction of the Stokes-shifted laser induced fluorescence (LIF) of the molecule is collected by the nanofiber, fiber-guided out of the cryostat, and monitored by a spectrometer (Shamrock SR-303i, Andor Technology) or two single photon counting modules (SPCMs). Contributions from the ex-

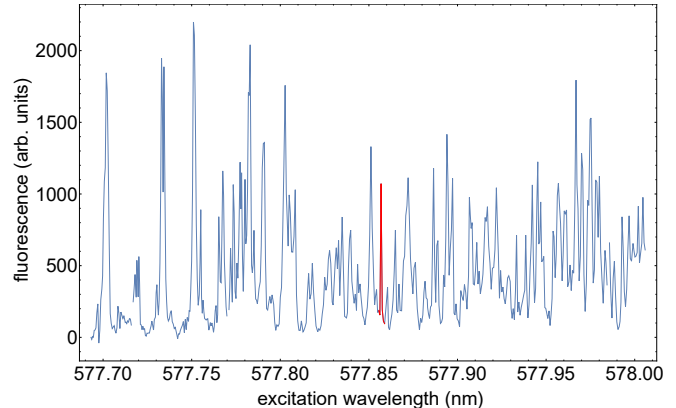


Figure 3. Fluorescence excitation measurement of nanofiber-interfaced terrylene molecules. The red peak indicates the fluorescence of a single terrylene molecule.

citation light and fluorescence on the 00ZPL are filtered out by a long-pass (LP) filter. Together with a short-pass (SP) filter to block Raman scattering from the fiber, this leaves a transmission window in the range of 630–650 nm. Due to the inhomogeneous line shifts induced by the host crystal matrix, single molecules can be spectrally selected with the narrowband dye laser if the terrylene concentration in the host crystal is small enough. Figure 3 shows the fluorescence excitation spectrum of a molecular ensemble in the X4 orientation. The excitation line of a verified single molecule is highlighted in red.

To characterize the optical interface created by individual molecules and the optical nanofiber, we have investigated four different single molecules that are located in the same nanocrystal. To verify the interaction with single quantum emitters, second order fluorescence intensity correlation measurements are performed by using a Hanbury-Brown-Twiss set-up [Fig. 1(a)]. The backscattered fluorescence is split by a 50/50 beamsplitter (BS) and directed to two SPCMs. Their counts are then recorded by means of a field programmable gate array and correlated. For a single quantum emitter, photon antibunching is observed, which manifests itself as an antibunching dip in the intensity correlation measurements at zero time delay τ between two detection events. Experimental imperfections such as incoherent background scatter will reduce the contrast of the dip but a dip deeper than 50% of the coincidence rates at long correlation times is proof of a single quantum emitter. Figure 4 shows second order fluorescence intensity

correlation measurements of a molecule on the nanofiber where the antibunching dip is clearly visible. As the exci-

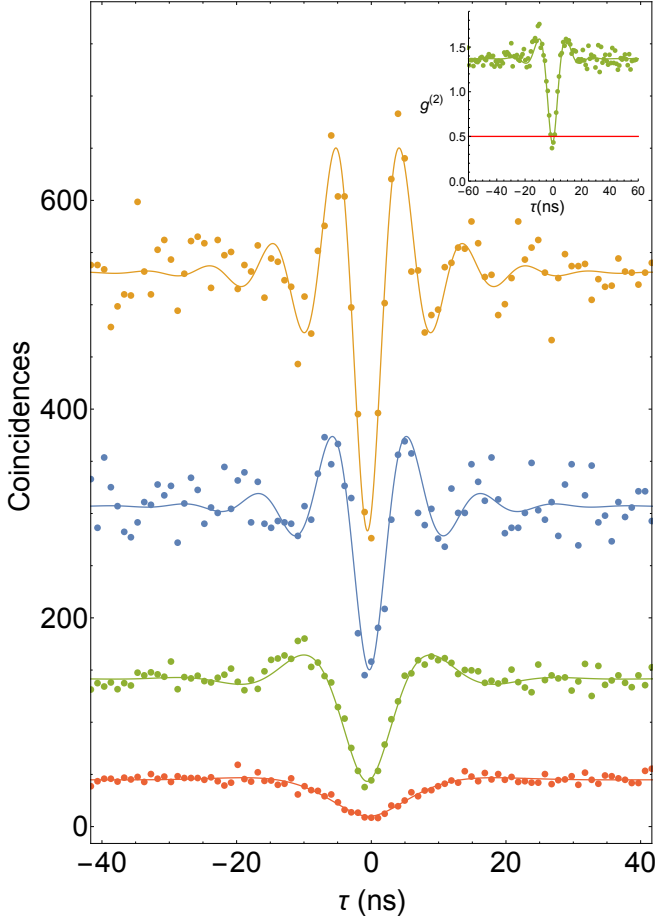


Figure 4. Fluorescence intensity correlation measurements of molecule C for different excitation powers (from bottom to top: 0.7 nW, 2.9 nW, 7.4 nW, 11.1 nW). The coincidences were recorded for 1000 s and the time resolution for the data is 1 ns. With increasing excitation power, the onset of Rabi oscillations and an increase in incoherent background scatter is clearly seen. The inset shows the $g^{(2)}$ -measurement at 2.9 nW normalised to the steady-state correlations of the fluorescence of the molecule, clearly indicating a single emitter.

tation intensity is increased, the onset of Rabi oscillations is apparent. Since the decay into the triplet state is negligible for short times, the measurements are fitted to the correlation function that is obtained by solving the optical Bloch equations for a two level system with a spontaneous decay rate Γ and a Rabi frequency $\Omega = \mathbf{d} \cdot \mathbf{E}/\hbar$, where \mathbf{d} is the dipole moment of the molecule and \mathbf{E} the electric field at the molecule's location [45].

$$g^{(2)}(\tau) = 1 - \left[\cos(\nu\tau) + \frac{3\Gamma}{4\nu} \sin(\nu\tau) \right] \exp\left(-\frac{3\Gamma\tau}{4}\right) \quad (2)$$

with

$$\nu = \sqrt{\Omega^2 - \left(\frac{\Gamma}{4}\right)^2}$$

An incoherent Poissonian background B added to a signal with average intensity $\langle I \rangle$ reduces the contrast of the intensity correlation measurements. This is taken into account when analyzing our data with the modified intensity correlation function [51]

$$g_B^{(2)}(\tau) = 1 + \frac{\langle I \rangle^2}{(\langle I \rangle + B)^2} (g^2(\tau) - 1). \quad (3)$$

We fit eqn.(3) to the data for different excitation powers while using Γ as a global fit parameter for all measurements on a given molecule. This yields the respective Rabi frequencies and the non-power broadened homogeneous linewidth Γ of the molecule. From these fits we also obtain the saturation intensity I_S of the molecule as $I/I_S = 2\Omega^2/\Gamma^2$. Figure 5 shows this expected linear increase of the squared Rabi frequency as a function of excitation power for three different molecules in the same crystal. The error bars obtained from fitting the intensity correlation measurements are smaller than the depicted datapoints in Fig. 5. From the fits, the saturation power P_S corresponding to I_S is obtained. We obtain saturation powers for molecule A of $P_S = 1.8 \pm 0.3$ nW, molecule B of $P_S = 4.3 \pm 0.8$ nW and molecule C of $P_S = 0.5 \pm 0.1$ nW. The biggest contribution to the error in the saturation powers arises from the uncertainty in determining the power in the fiber. We convert P_S to the maximum intensity at the surface of the nanofiber. Without the exact knowledge of the orientation of the molecule's transition dipole moment and its distance from the nanofiber surface, this gives an upper limit for the saturation intensity. The measured saturation intensities are $I_S < 1.8$ Wcm $^{-2}$ for molecule A, $I_S < 4.3$ Wcm $^{-2}$ for molecule B and $I_S < 0.5$ Wcm $^{-2}$ for molecule C. These results compare well with results obtained by other groups who studied terrylene in bulk p-terphenyl [37, 52]. To our knowledge a saturation intensity of $I_S < 0.5$ Wcm $^{-2}$ as for molecule C is the lowest measured so far for terrylene in p-terphenyl. As opposed to measurements on terrylene in bulk p-terphenyl using a confocal microscope, the excitation light in our case enters through the side of the thin host crystal platelets. As the transition dipole moment of the molecules lies nearly perpendicular to the base of these platelets, this suggests an improved overlap between the polarization of the nanofiber-guided excitation light and the transition dipole moment of the molecules. An independent measurement of the saturation power is obtained by recording the resonant fluorescence rate R_{LIF} as a function of excitation power as plotted in Fig. 6, which includes a fit to $R_{\text{LIF}} = R_\infty \frac{P/P_S}{1+P/P_S}$. The error bars on the fluorescence rate that are obtained by taking the standard error of the amplitude from fits over several molecular spectra are smaller than the depicted datapoints. This measurement yields a saturation power of 4.8 ± 1.4 nW for molecule B, where the error stems from the fit and from the uncertainty in the excitation power inside the fiber. This translates into a

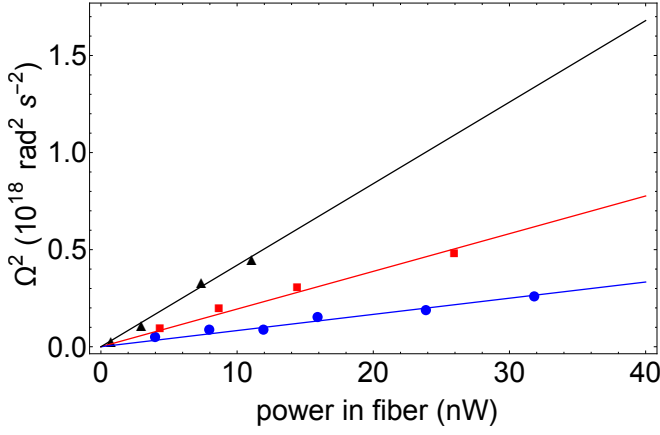


Figure 5. Squared Rabi frequency as a function of excitation power for molecule A (red, squares), B (blue, circles) and C (black, triangles) and corresponding linear fits.

saturation intensity of $I_S < 4.8 \text{ Wcm}^{-2}$ for molecule B in good agreement with $I_S < 4.3 \text{ Wcm}^{-2}$ as obtained with the HBT setup. We performed a further measure-

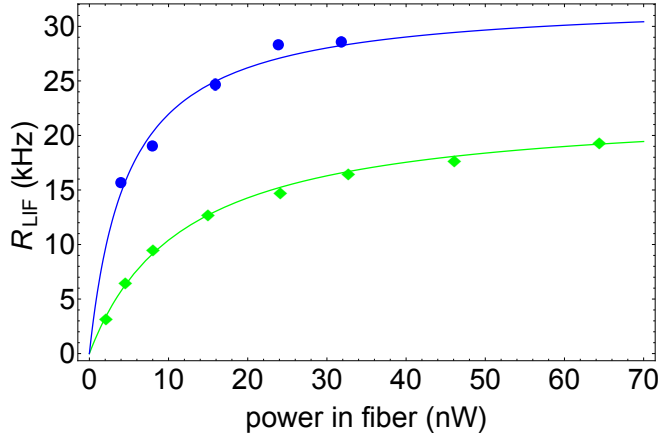


Figure 6. Saturation of the resonant fluorescence intensity R_{LIF} as the excitation laser power is increased for molecules B (blue, circles) and D (green, diamonds) and corresponding fits.

ment on a fourth molecule (molecule D) that yielded a lower fluorescence rate and indeed was measured to have a higher saturation power of $11.4 \pm 2.2 \text{ nW}$ and therefore a saturation intensity of $< 11.4 \text{ Wcm}^{-2}$. This suggests that this molecule is located further from the nanofiber surface such that its fluorescence does not couple back to the nanofiber-guided modes as efficiently. Alternatively, the alignment between polarization of the excitation light and the dipole moment of molecule D may be less favorable. Although all molecules are embedded in a single crystal and therefore have the same orientation, the inherent birefringence of the host crystal can cause a less favorable alignment: The phase shift between the corresponding polarization components is $\Delta\phi = 2\pi\Delta nL/\lambda$,

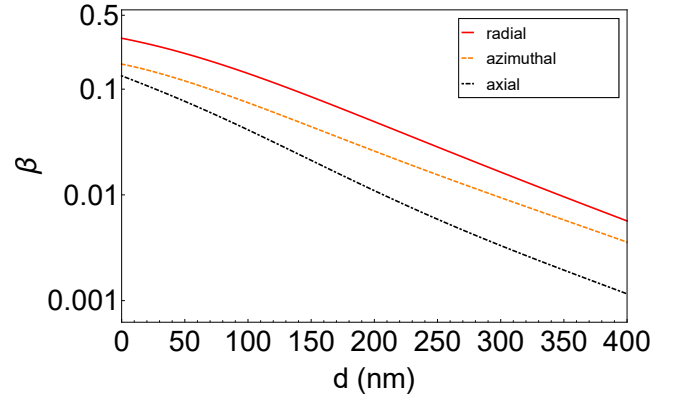


Figure 7. Mean coupling efficiency of a linear radial (red), tangential (orange, dashed) and axial (black, dot-dashed) dipole, radiating at 630–650 nm, to the nanofiber as a function of distance from the nanofiber surface.

where λ is the vacuum wavelength, L the propagation distance and Δn is the effective birefringence that can reach 0.32 in our case. The overall efficiency for fluorescence excitation and detection via the nanofiber interface is given as $\eta_{\text{LIF}} \propto |\mathbf{d} \cdot \mathbf{E}|^2 \beta$, where $\beta = \Gamma_g/\Gamma_{sc}$ is the coupling efficiency of dipole radiation to the nanofiber modes. This coupling efficiency depends on the distance and the orientation of the dipole with respect to the nanofiber surface. Here, Γ_g is the scattering rate into guided modes and Γ_{sc} is the total scattering rate of the dipole. For our case of a nanofiber with 160 nm radius and Stokes shifted fluorescence in the range of 630–650 nm, we calculated this coupling efficiency for a radially, azimuthally and axially oriented dipole following [33], see Fig. 7.

Since the saturation intensity measured for molecule C is the lowest yet measured for terrylene in p-terphenyl, we assume it to be very close to the surface of the nanofiber. An upper limit for the other molecules from the nanofiber surface can then be estimated by comparing their saturation intensities. Because the host crystal is birefringent, we only give an upper limit on the radial distance between the different molecules. Figure 8 shows the calculated efficiency of fluorescence excitation and detection via the nanofiber interface for different positions of the dipole with respect to the nanofiber surface. The positions are chosen to lie within the volume of a platelet crystal with its base on the nanofiber surface. The molecular dipole is oriented perpendicular to the crystal's base and excited by quasilinear light via the nanofiber-based interface. Assuming unpolarised light instead of quasilinear polarised light has a negligible effect of less than 3% on the relative distances between the different molecules. We did not incorporate the refractive indices of the crystal into this model because they would make the local efficiencies very dependent on the crystal's geometry and we are only interested in assigning the maximum radial distance of the measured molecules. The maximum ra-

dial distances of the investigated molecules are depicted in their respective colours by dashed contours [Fig. 8]. These results show that they radially all lie within less than 280 nm of each other and less than 160 nm from the nanofiber surface. This translates into coupling efficiencies for a radial dipole to the nanofiber mode between 8-30% [Fig. 7].

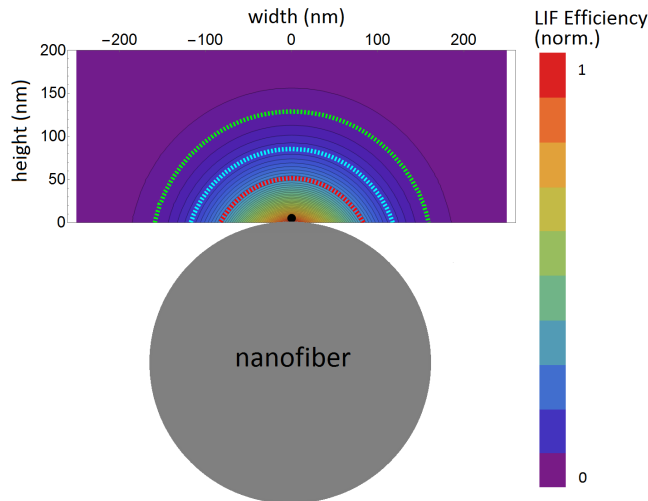


Figure 8. The excitation and detection efficiency η_{LIF} for a molecular dipole at different positions with respect to the nanofiber. The spatial coordinates are chosen assuming a platelet crystal that is lying with its base on the nanofiber surface. The contact point of nanofiber and crystal is at the origin of this reference frame. The efficiency is normalised to the maximum efficiency on the nanofiber surface of 8% (excitation and detection). The maximum radial distances for the different molecules are indicated by dashed colored lines (A = red, B = blue, D = green, from bottom to top). The reference molecule C is shown as a black dot and is assumed to be located directly on the nanofiber surface at position $(x,y) = (0,0)$.

Summarizing, we have shown how single molecules can be optically interfaced via the evanescent field surrounding an optical nanofiber. This is an important addition to the toolbox of quantum emitters such as atoms [31, 53, 54], quantum dots [30, 55] and NV centers in nanodiamonds [36, 56] that have been fiber-integrated by coupling to optical nanofibers. Each of these systems has its own intrinsic advantages for their usage in quantum networks. Single molecules in solids are efficient quantum emitters that can exhibit a broad range of emission wavelengths. This makes them suitable to be interfaced with other quantum emitters [1]. They have an advantageous level structure for the implementation of triggered single photon sources [38, 57] and have proven their versatility in quantum optics [58–60]. Further, single waveguide-coupled molecules allow the investigation of photon-mediated interactions between two quantum emitters even when they are separated by much more

than the excitation wavelength [61–63]. These interactions can be further enhanced by using a nanofiber between two fiber Bragg gratings and thereby realizing a high-Q cavity [64]. Single molecules that are coupled to the evanescent field of optical nanofibers therefore not only offer a rich experimental platform for investigating entanglement and correlations between quantum emitters, they also provide a means for implementing components of quantum networks such as fiber-coupled single photon sources [29, 65, 66] or photon sorters [67, 68].

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